

The normal state of URu₂Si₂: spectroscopic evidence for an anomalous Fermi liquid.

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We report on new measurements of the optical conductivity of the heavy fermion compound URu₂Si₂ using a new technique that reduces the noise level of the spectra by an order of magnitude. In the normal state, above the hidden order transition at 17.5 K, we find that the optical resistivity, defined as $\rho(\omega) = \text{Re}(1/\sigma(\omega))$ can be accurately described by a generalized Fermi liquid like formula where $\rho(\omega, T) = A'(\omega^2 + b(\pi T)^2) + \rho_0$. The optical measurement is able to establish both A' and b and can be compared with dc transport that yields $A'b$. We find that instead of the value of $b = 4$ expected for a Fermi liquid, our experiments show that $b \approx 1$. This value of b is a signature of *elastic scattering*. Since the scattering is intrinsic we discuss this normal state in terms of an unconventional incoherent Fermi liquid, Fermi molasses.

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It was shown by Landau that electrons in a metal obey Fermi statistics and continue to act like free quantum mechanical particles but with enhanced masses[1]. This state of matter, the Landau Fermi liquid, is recognized experimentally by an electrical resistivity that is proportional T^2 plus a term proportional to the square of the frequency of the applied field. The ratio of the two terms can be expressed by a dimensionless constant b . Calculations show that $b = 4$ in a Landau Fermi liquid[2]. In conventional metals, Fermi liquid effects are weak and are masked by the electron-phonon interaction. In the heavy Fermion materials Fermi liquid scattering dominates and among these URu₂Si₂ is a good model system where the enigmatic hidden order transition at 17.5 K has generated much attention as well as new spectroscopic data[3–5]. We decided to do a detailed study of the Fermi liquid effects of this system using infrared spectroscopy with a focus on the normal state above the hidden order transition. Fermi liquid like states with $b = 1$ have been observed in a number of disparate systems[6–10] but the significance of this result has not been recognized.

Among the heavy Fermion metals URu₂Si₂ is one of the most interesting. It displays, in succession, no less than four different behaviours. At room temperature it has a Kondo-like resistivity, rising slightly as the temperature is lowered as the conduction electrons are incoherently scattered by localized uranium f electrons. Below $T_K \approx 75$ K, the resistivity begins to drop and the material resembles a typical heavy Fermion metal[11, 12]. Below $T_0 = 17.5$ K the hidden order phase transition gaps a substantial portion of the Fermi surface but the nature of the order parameter is not known. Finally, at 1.5 K URu₂Si₂ becomes an unconventional supercon-

ductor. Experiments of Schmidt *et al.* [5] find a light band crossing the Fermi surface above 17.5 K but turning into a heavy $m^*/m_e = 25$ band only at the hidden order transition. This contradicts the conventional view that mass builds up gradually below T_K . We can test this by carefully tracking the Drude weight as a function of temperature with optical spectroscopy. The early optical experiments of Bonn *et al.* [13] showed that URu₂Si₂ at 20 K, above the hidden order transition, has an infrared spectrum consisting of a narrow Drude peak and a strong incoherent background. This Drude weight is a quantitative measure of the effective mass of the carriers.

The single crystals of URu₂Si₂ were grown at Grenoble and at McMaster in tri-arc furnaces in an argon atmosphere. The crystals were annealed, under UHV, at 900 C for 10 days. The ab plane cleaved surfaces were measured by standard reflectance techniques, at three separate laboratories, using an *in situ* gold overcoating technique[14]. The absolute reflectance results of the three groups agreed to within 0.5 %. At long wavelengths a simple procedure which we call “refined thermal reflectance” was used to improve the signal to noise ratio. The reflected signal is measured over a relatively narrow range of temperatures without any physical motion of the sample. Then a gold overcoating is applied and the absolute reflectance is calculated. Next we fit a cubic polynomial to one of the absolute spectra to produce a smooth curve averaging out the artifacts. This smoothed spectrum is then used as a reference spectrum for all the other temperature dependent spectra. The measured refined reflectance was converted to an optical conductivity by Kramers-Kronig analysis. At low frequency, below 2 meV, a Drude response was assumed where we used the

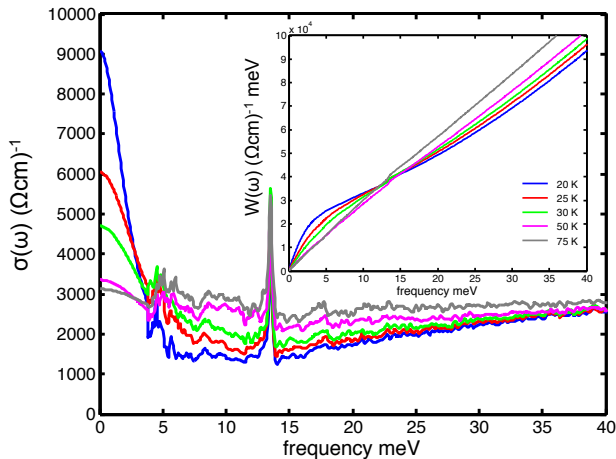


FIG. 1: (color online): The optical conductivity as a function of photon energy in the heavy Fermion state. Below 4 meV the conductivity has been fitted to a Drude peak whose amplitude agrees with the dc resistivity. The inset shows the accumulated spectral weight up to a frequency ω . Below 10 meV the Drude weight dominates. The total Drude weight is temperature independent since all the curves join at 15 meV above the Drude cutoff. The high frequency spectral weight is not conserved since the spectral weight in the hybridization gap region, 5 to 40 meV, is lost to higher frequencies and the cumulated spectral weight drops at 40 meV as the temperature is lowered.

measured dc resistivity to determine the amplitude of the Drude peak and the absorption at our lowest measured infrared frequency to determine the width. At high frequency, beyond 7 eV, we used the results of Degiorgi *et al.* [15].

Figure 1 shows the optical conductivity between 20 and 75 K, the region where coherence develops as shown by the appearance of a Drude peak below 15 meV which narrows as the temperature is lowered. Above 75 K the optical conductivity is frequency and temperature independent. Interestingly, we find that in the temperature range 75 K to 20 K the area under the Drude peak is temperature independent with a plasma frequency of ≈ 400 meV, a signature that m^* is constant in this region of temperatures and a distinct minimum develops between the Drude peak and the high frequency saturation value. We suggest this minimum is a pseudo-hybridization gap normally associated with the formation of the Kondo lattice but not fully formed in this material above 17.5 K. There is a simple relationship between the Kondo temperature T_K , the effective mass m^* and the gap V_K : $m^*/m_e = (V_K/k_B T_K)^2$ [16, 17]. Estimating $T_K = 75$ K from the temperature where the Drude peak first appears, and taking $V_K = 15 \pm 5$ meV, we find $m^*/m_e = 5 \pm 2$ which is lower than what is estimated from specific heat measurements[12] but not in disagreement with recent STM data[5]. We note here that the

hybridization gap acts like the pseudogap in the cuprates. Its frequency does not change with temperature but fills in gradually as the temperature is raised. Also, the spectral weight lost in the gap region is not recovered by the Drude peak or in the spectral region immediately above the gap. The inset shows the accumulated spectral weight at the five temperatures. All the curves cross at 15 meV showing that the Drude weight is conserved in the temperature range from 20 to 75 K. On the other hand, spectral weight is lost above this frequency range as the temperature is lowered. These behaviours are inconsistent with a simple picture of an effective mass resulting from an inelastic interaction with a bosonic spectrum.

To examine quasiparticle damping above the hidden order transition we apply an extended Drude model to the conductivity:

$$\sigma(T, \omega) = \frac{\Omega_p^2}{4\pi} \frac{1}{1/\tau^{op}(\omega) - i\omega(1 + \lambda(\omega))} \quad (1)$$

where $\Omega_p^2 = 4\pi n e^2/m$ is the plasma frequency squared, $1/\tau^{op}(\omega) = \frac{\Omega_p^2}{4\pi} \mathcal{R}e(1/\sigma(\omega))$, the optical scattering rate, and $1 + \lambda(\omega) = m^*/m$ is the mass enhancement. Optical phonons at 13.5 and 46.9 meV have been subtracted from the measured conductivity. The optical scattering rate is shown in figure 2 a) where we have used a plasma frequency of $\Omega_p = 418$ meV, evaluated from the Drude weight. As temperature exceeds T_K , here taken as 75 K, the frequency dependence below 14 meV is replaced by uniform temperature and frequency independent scattering. We also note that the scattering above 20 K is *incoherent* in the sense that $1/\tau > \omega$ but, significantly, the condition reverses at 20 K, near the temperature of the hidden order transition.

The choice of plasma frequency is to some extent uncertain and to better compare the scattering rate with transport data we turn to a directly measured quantity, the optical resistivity, defined as $\rho(\omega) = \mathcal{R}e(1/\sigma(\omega))$ where $\sigma(\omega)$ is the complex conductivity. This quantity is plotted in figure 2 b) at three temperatures. The zero frequency limit of $\rho(\omega)$ is the dc resistivity, which, as mentioned above, has been adjusted to agree with the measured resistivity. The filled circles at zero frequency show the measured dc resistivity. Figure 2 b) also shows a parabola fitted to the data where the constant A' and a dc offset ρ_0 are adjustable parameters. The offset is related to residual elastic scattering. Also contributing to the offset are experimental uncertainties at the 0.3 % level in the absolute reflectance. We will now turn to discuss this model. We have also added a term $A'(\pi T)^2$ to the offset to account for the temperature term in Fermi liquid scattering (see below).

It is generally assumed that a quadratic frequency (and temperature) dependence of the scattering rate is a signature of Fermi liquid scattering. The frequency dependent resistivity is then given by: $\rho(\omega, T) = A'(\omega^2 +$

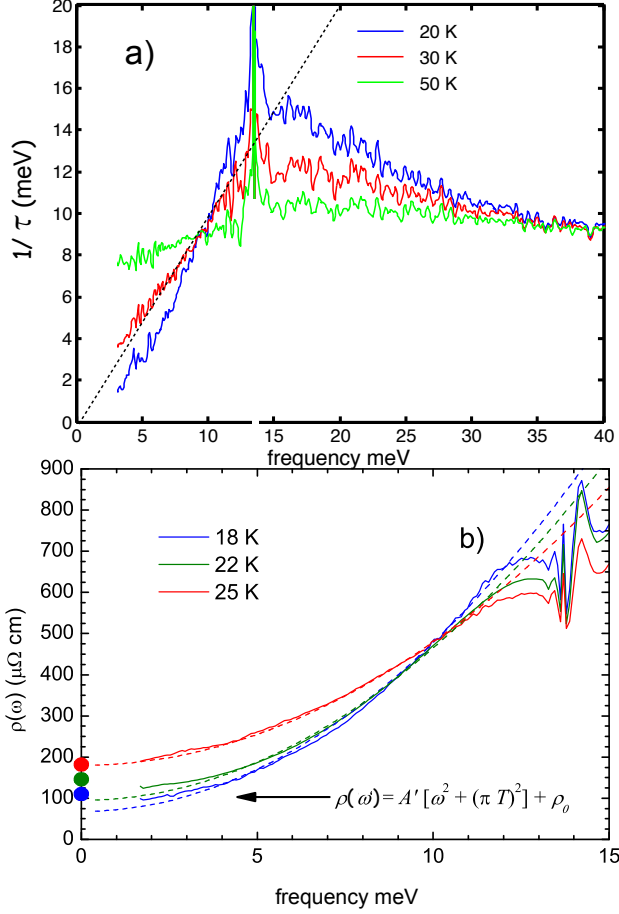


FIG. 2: (color online). a) The frequency dependent scattering rate at three temperatures in the normal state above the hidden order transition at 17.5 K. As the temperature is raised the Fermi liquid scattering below 14 meV is replaced by a uniform frequency independent incoherent scattering. Coherent quasiparticles exist below the dashed line $\omega > 1/\tau$. b) The optical resistivity $\rho(\omega)$ vs. photon energy at low frequencies. The experimental curves (solid lines) are compared to a Fermi liquid fit (dashed lines) with the coefficient A' and an offset ρ_0 determined by a least squares fit to the experimental data. From 18 to 22 to 25 K, values of $A'\pi^2$ decrease from 0.3 to 0.27 to 0.22 $\mu\Omega \text{ cmK}^2$.

$(2\pi T)^2$ [2, 16]. To be slightly more general we introduce a second parameter b to allow the ratio between the temperature and frequency terms to vary from the predicted Fermi liquid value where $b = 4$. Then the frequency dependent resistivity is given by $\rho(\omega, T) = A'(\omega^2 + b(\pi T)^2) + \rho_0$ where ρ_0 is the residual resistivity. The parameter A' can be related to the commonly used Fermi liquid parameter A , as it is defined in dc resistivity measurements, $\rho = AT^2$. With these definitions b is given by $b = A/(A'\pi^2)$. We can determine A' directly from a quadratic fit to the optical data shown in figure 2 b). Note that the scattering rate deviates from the simple quadratic form below 5 meV and above 12 meV

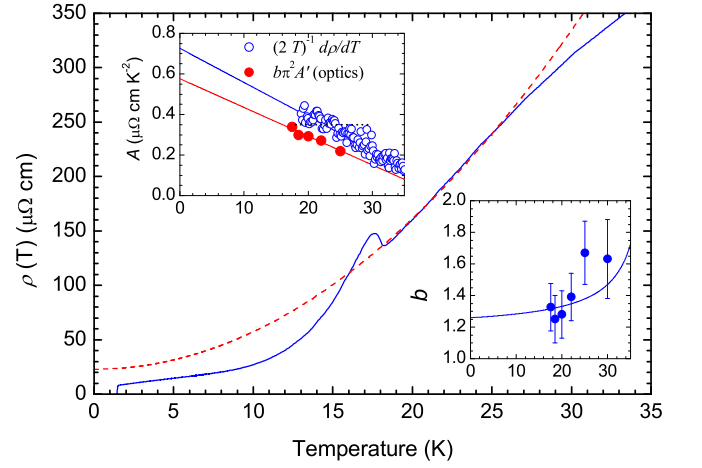


FIG. 3: (color online). The solid line in the main panel shows the experimental dc resistivity of URu₂Si₂ ref.[19]. The dashed line is a parabolic fit ($\rho(T) = \rho_0 + AT^2$ with $\rho_0 = 23\mu\Omega \text{ cm}$ and $A = 0.35\mu\Omega \text{ cmK}^2$ in the temperature range 18–27 K. The top inset shows the temperature evolution of the parameter A obtained from the derivative of the dc resistivity and from parabolic fits to the optical resistivity. They have an approximate linear temperature dependence shown as solid lines. The dotted line is A from the parabolic fit to $\rho(T)$. The bottom inset shows the parameter b as a function of temperature. The solid line is the ratio of the straight lines shown in the top inset.

where it saturates. As the temperature increases the coefficient A' decreases whereas the cutoff seems to remain at 12 meV. Even with our enhanced signal to noise ratio we see little evidence to coupling to sharp resonance modes in our spectra of the type seen in the cuprates[18]. *The self energy of the quasiparticles is dominated by a featureless continuum without an energy scale.*

We expect the dc resistivity to vary as AT^2 and, as shown in figure 3, we do get a reasonable fit in the 18 to 27 K temperature range of the optical data without a linear term and a residual resistivity that agrees with the dc resistivity data. However, in our analysis we use the temperature derivative from Zhu *et al.* [19] to determine the dc resistivity coefficient A . From the optically determined A' and the dc resistivity value A we can determine the ratio b as a function of temperature, which is shown in the inset to figure 3. Instead of the expected value of $b = 4$ for Fermi liquid scattering[2] our data clearly show that $b \approx 1$ in the temperature region immediately above the hidden order transition. This discrepancy is well outside our possible error. There is a trend for b to increase with temperature while there is a much weaker trend in the opposite direction for A . The value $b = 1$ is expected for resonant elastic scattering from impurities[20], when the single-particle scattering rate has an ω^2 but no T^2 term. The Kubo formula then yields the optical $1/\tau$ with $b = 1$. Here, however, the scattering appears to be intrinsic. One possibility is that in this material, in-

stead of the formation of an Anderson lattice of coherent states, the uranium f levels act like independent incoherent scatterers and form the coherent Kondo lattice only below the hidden order transition. This picture has also been advanced by Haule and Kotliar[21] and Schmidt *et al.* [5] Our data provide independent evidence for this model. An anomalously low ratio of temperature to the frequency dependence has also been observed in UPt_3 by Sulewski *et al.* [6] where an upper limit on $b = 1$ was estimated. The infrared data of Basov *et al.* [9] on chromium and Katsufuji *et al.* on Sr_2RuO_4 [7] are also consistent with $b \approx 1$. Fermi liquid behaviour with $b = 1$ has also been reported in $\text{Ce}_{0.95}\text{Ca}_{0.05}\text{TiO}_{3.04}$ by Katsufuji and Tokura[8] and in $\text{Nd}_{1-x}\text{TiO}_3$ by Yang *et al.* [10]. Another comparison between the temperature and frequency dependence of scattering is the ratio of the Kondo temperature $T_K = 75$ K and the cutoff frequency $\omega_c = 14$ meV of frequency dependent scattering. If it is written as $b_c = \omega_c^2/\pi^2 T_K^2$ we find that $b_c = 0.48$, again substantially smaller than the Fermi liquid value of $b = 4$. We see from figure 2 a) that the scattering rate exceeds the frequency, *i.e.* $1/\tau > \omega$ at all frequencies above 2 meV *i.e.* the quasiparticles are incoherent. The state of matter above the hidden order transition seems to obey Fermi statistics in that it shows a quadratic frequency and temperature dependence of the scattering rate and yet does not show well-defined coherent quasiparticle states as expected for a true Landau-Fermi liquid. We call this state Fermi molasses, a sticky Fermi liquid. The important question remains, are there cases of true Fermi liquids with $b = 4$? The only example we have found is $b = 5.6$, reported by Dressel[22] in the organic quasi-2D compound $\kappa\text{-(BEDT-TTF)}_2\text{Cu[N(CN)}_2\text{]Br}_x\text{Cl}_{1-x}$.

In summary, we have found that in the normal state above the hidden order transition a relatively light band with a mass $m^*/m_e \approx 5$ is weakly coupled to the f electrons with $V_K \approx 5$ meV, and that this band is responsible for the transport current as measured by the optical conductivity. We suggest that this coupling is not strong enough to form an Anderson lattice. Instead the f electrons act like elastic, incoherent scatterers as shown by the anomalous $b = 1$ in the generalized Fermi liquid scattering formula instead of the expected $b = 4$ for coherent inelastic scattering from bosonic excitations. As suggested by the STM experiments of Schmidt *et al.* the Fermi liquid with the heavy quasiparticles exists only below the hidden order transition. Because of the rapidly varying electronic density of states we are unable to use our technique to analyze the nature of the scattering below the hidden order transition to verify this scenario, but if accurate experiments were possible, we would expect the recovery of $b = 4$ behaviour at low temperature, as expected for a genuine Fermi liquid. We also note that this anomalous Fermi liquid behaviour is shared by a number of other strongly correlated materials where magnetism appears to play a role.

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